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LETTER TO THE EDITOR

The formation and magnetic properties of $Dy_2Fe_{17}C_x$ with high carbon content obtained by rapid solidification

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Abstract. The carbides $Dy_2Fe_{17}C_x$ with x = 1.5–3.0 have been successfully prepared by the melt-spinning method. The x-ray diffraction patterns and the thermomagnetic curves show that they are of single phase in general with Th_2Zn_{17} structure type when x = 1.5, 2.0, 2.5 and 2.8 with a small amount of α-Fe as an impurity phase, while for x = 3.0 the proportion of the α-Fe phase is high. The lattice parameters a and c are both enlarged and increase with the carbon content x. The Curie temperatures T_c are greatly enhanced when $x < 2.5$ and remain almost constant when x = 2.5–3.0, but the saturation magnetizations, σ_s, increase slightly. The $Dy_2Fe_{17}C_x$ ribbons still maintain the rhombohedral Th_2Zn_{17}-type structure after being annealed at 1100 °C for 14 h, which indicates the high stability of $Dy_2Fe_{17}C_x$ (x = 1.5–3.0) compounds obtained by rapid solidification. The heat-treated ribbon samples can be magnetically oriented and prefer an easy-plane anisotropy at room temperature.

Early in 1988 [1], it was found that $R_2Fe_{14}C$ decomposed into stable ternary carbides, $R_2Fe_{17}C_x$, with the rhombohedral Th_2Zn_{17} or the hexagonal Th_2Ni_{17} structure type. Neutron diffraction data showed that the carbon atoms interstitially occupied part of the empty 9(e) site [2]. Tm and Gd Mössbauer spectroscopies on $R_2Fe_{17}C_x$ suggested a large influence on the crystalline electric field experienced by the 4f electrons of the rare earth atoms upon carbonation [3, 4]. An easy-axis anisotropy field of 5.3 T at room temperature had been achieved in the $Sm_2Fe_{17}C$ alloy [5]. Further efforts to obtain $R_2Fe_{17}C_x$ compounds by melting led to a maximum occupancy around x = 1 [5, 6]. Recently through solid–gas reaction, higher carbon contents up to x = 2 have been obtained, which resulted in a T_c of about 400 °C and a high uniaxial anisotropy field of more than 10 T for $Sm_2Fe_{17}C_2$ [7, 8]. By mechanical alloying and a subsequent solid–gas reaction in acetylene, Kuhrt *et al* [9] achieved a room temperature coercivity of up to 18.5 kA cm^{-1} (23.2 kOe) and a maximum energy product of 59 kJ m^{-3} (7.4 MG Oe) for the microcrystalline $Sm_2Fe_{17}C_2$ powders. Hence, the $R_2Fe_{17}C_x$ series with high carbon concentration still attracts great interest both for application and basic research purposes. A big problem of the $R_2Fe_{17}C_x$ series carbided by a solid–gas reaction is their easy decomposition into RC and α-Fe [9], which is a hindrance with regard to permanent magnet applications.

Previously, we have succeeded in preparing the carbides $R_2Fe_{17}C_x$ with high carbon content by the melt-spinning method. Here we study the formation, the phase stability and the magnetic properties of the series $Dy_2Fe_{17}C_x$ (x = 1.5, 2.0, 2.5, 2.8, 3.0) prepared by rapid solidification.

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Alloys were made first by arc melting Fe and C to form Fe–C alloys, and then melting several times with iron and rare earth elements in the series $Dy_2Fe_{17}C_x$ with $x = 1.5$, 2.0, 2.5, 2.8 and 3.0. Both meltings were under purified argon atmospheres and the elements used were at least 99.9% pure. The homogeneous ingots of about 3 g were finally rapidly quenched in a purified argon atmosphere by ejecting the molten alloy onto the surface of a rotating copper-wheel roller through a nozzle at the bottom of a quartz tube. The range of the surface velocity V_s of the copper disk is from 0–47 m s^{-1}. The ribbons obtained were about 20 μm thick and 1.5 mm wide. Some of the melt-spun ribbons were annealed at 1100 °C for 14 h, while under a pressure of 5×10^{-5} Torr. The finely ground samples were oriented in an applied field of 10 kOe in an epoxy resin.

X-ray diffraction was performed on powder samples using Co Kα radiation to determine the phase structure. The Curie temperatures were derived from the temperature dependence of the magnetization $\sigma(T)$ curves measured by a vibrating sample magnetometer in a field of 2 kOe. The magnetizations at 1.5 K and 293 K were obtained from magnetization curves measured in a field up to 70 kOe by an extracting sample magnetometer.

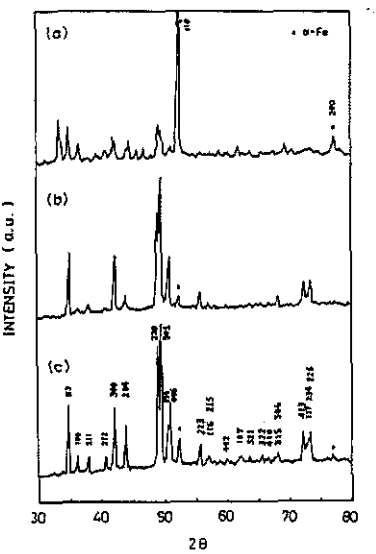

Figure 1. X-ray diffraction diagram of the $Dy_2Fe_{17}C_{2.0}$ compound with Co Kα radiation: (*a*) the arc-melted alloy; (*b*) the melt-spun ribbon at $V_s = 16$ m s^{-1}; (*c*) the ribbon heat treated at 1100 °C for 14 h.

X-ray diffraction indicates that at certain quenching rates, with V_s changing for different carbon content x, the $Dy_2Fe_{17}C_x$ ($1.5 < x < 3.0$) compounds crystallize in the rhombohedral Th_2Zn_{17}-type structure. A typical example is shown in figure 1. In arc-melted alloys $Dy_2Fe_{17}C_x$ ($x > 1.5$) the α-Fe is the majority phase together with DyC compounds (figure 1(*a*)). Gradually raising the quenching rate, the amount of α-Fe is reduced until it forms a small impurity phase and the Th_2Zn_{17}-type structure becomes dominant (figure 1(*b*)). For $Dy_2Fe_{17}C_{2.0}$ the appropriate V_s is

about 16 m s^{-1}. There is still a large amount of α-Fe in the $Dy_2Fe_{17}C_{3.0}$ ribbon samples and, on raising the quenching rate, the higher V_s value leads to the formation of the amorphous phase.

Table 1. Lattice parameters a and c, unit cell volume V, relative change of unit cell volume $\Delta V/V$ upon carbonation, Curie temperature T_c and saturation magnetization σ_s at T = 1.5 K and T = 293 K for the $Dy_2Fe_{17}C_x$ ribbons.

Compound	a (Å)	c (Å)	V (Å^3)	$\Delta V/V$ (%)	T_c (K)	σ_s (emu g^{-1})	
						T = 1.5 K	T = 293 K
Dy_2Fe_{17} [a]	8.767	8.312	516				
$Dy_2Fe_{17}C_{1.0}$ [b]	8.585	12.454	794.9	2.7	515		73.78
$Dy_2Fe_{17}C_{1.5}$	8.621	12.499	804.6	4.0	578	66.31	79.70
$Dy_2Fe_{17}C_{2.0}$	8.645	12.585	814.6	5.2	626	69.13	84.28
$Dy_2Fe_{17}C_{2.5}$	8.669	12.599	820.1	6.0	680	64.22	88.46
$Dy_2Fe_{17}C_{2.8}$	8.677	12.613	822.5	6.3	676	72.46	92.60
$Dy_2Fe_{17}C_{3.0}$	8.695	12.600	825.0	6.6	681		

[a] Values obtained from [11].
[b] Values obtained from [6].

The lattice parameters a and c, the unit cell volume V and the relative change of the unit cell volume $\Delta V/V$ are given in table 1. The values of the parent Dy_2Fe_{17} [11] and the $Dy_2Fe_{17}C$ [6] alloys are also given in table 1 for comparison. Both lattice parameters a and c, and the unit cell volume V dilate upon carbonation and increase with the carbon concentration x. This lattice expansion is consistent with the previous results [6] as shown in figure 2. The relative change of the unit cell volume $\Delta V/V$ is 4.0–6.6%, which is comparable to that of the corresponding nitrides of R_2Fe_{17} [11]. The transition from the hexagonal structure of Dy_2Fe_{17} to the rhombohedral structure of $Dy_2Fe_{17}C_{1.5}$ takes place at lower carbon content within $0 < x < 1.5$, as for the other series of $R_2Fe_{17}C_x$ [12].

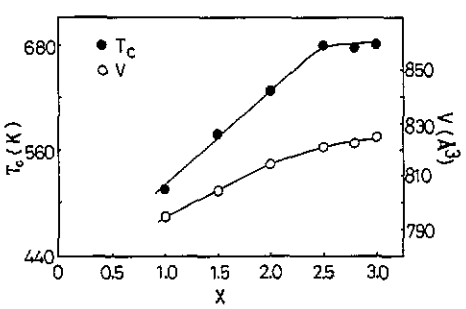

Figure 2. The Curie temperature T_c and the unit cell volume V as functions of the carbon content x of the $Dy_2Fe_{17}C_x$ series.

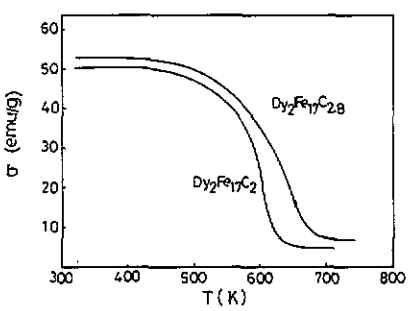

Figure 3. The temperature dependence of the magnetization of $Dy_2Fe_{17}C_{2.0}$ and $Dy_2Fe_{17}C_{2.8}$.

The x-ray diffraction pattern figure 1(*c*) shows that the $Dy_2Fe_{17}C_{2.0}$ ribbons still maintain the Th_2Zn_{17}-type structure after being heat treated at 1100 °C for 14 h, demonstrating the high stability of the ternary carbides $R_2Fe_{17}C_x$ with high carbon

content prepared by the melt-spinning method, in contrast to the metastability of the carbides $R_2Fe_{17}C_x$ prepared by solid–gas reaction. The $Dy_2Fe_{17}C_x$ ribbons can be oriented after heat treatment and prefer an easy-plane magnetization anisotropy.

Figure 3 shows the temperature dependence of the magnetization of the $Dy_2Fe_{17}C_{2.0}$ and $Dy_2Fe_{17}C_{2.8}$ samples. All these samples are approximately single phase. The saturation magnetization σ_s at $T = 1.5$ K and $T = 293$ K and the Curie temperature T_c are also summarized in table 1. $\sigma_s(1.5\ \text{K})$ and $\sigma_s(293\ \text{K})$ increase slightly with the carbon concentration x, except for in the case of $Dy_2Fe_{17}C_{3.0}$ due to its high content of α-Fe. The dependence of the Curie temperature on carbon content x is shown in figure 2. It can be seen that the increase of C uptake produces a marked increase in the Curie temperature, due to the enhancement of the Fe–Fe exchange interaction by the interstitial carbon atoms. There is little change in the Curie temperature when $x > 2.5$ in spite of the slight increase of the unit cell volume.

In summary, we have made the highly stable ternary carbides $Dy_2Fe_{17}C_x$ with high carbon content ($x > 2$) by the melt-spinning method. The Curie temperature increases considerably and the saturation magnetization increases slightly accompanied by a dilation of the unit cell volume.

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